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Julian B. Andelman

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16. ABSTRACT

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Using a simplified indoor air model and assuming complete volatilization from a full range of typical water uses within the home, calculations indicate that the expected air inhalation exposures can be substantially higher than those from ingestion of these chemicals in drinking water. Although the regulation of toxic chemicals in potable water supplies has focused traditionally on direct ingestion, the volatilization and inhalation from other much greater volume indoor uses of water should be considered as well.

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Human Exposures to Volatile Halogenated Organic Chemicals in Indoor and Outdoor Air

by Julian B. Andelman*

Volatile halogenated organic chemicals are found in indoor and outdoor air, often at concentrations substantially above those in remote, unpopulated areas. The outdoor ambient concentrations vary considerably among sampling stations throughout the United States, as well as diurnally and daily. The vapor pressures and air–water equilibrium (Henry's Law) constants of these chemicals influence considerably the likely relative human exposures for the air and water routes.

Volatilization of chemicals from indoor uses of water can be a substantial source of exposure, as shown for radon-222. Measurements of air concentrations of trichloroethylene (TCE) in showers using TCE contaminated groundwater show increases with time to as high as one-third of occupational threshold limit values. Using a scaled down experimental shower, such volatilization and subsequent decay in air was also demonstrated.

Using a simplified indoor air model and assuming complete volatilization from a full range of typical water uses within the home, calculations indicate that the expected air inhalation exposures can be substantially higher than those from ingestion of these chemicals in drinking water. Although the regulation of toxic chemicals in potable water supplies has focused traditionally on direct ingestion, the volatilization and inhalation from other much greater volume indoor uses of water should be considered as well.

Introduction

As measurement capabilities increase along with awareness and concern about the releases to the air of a variety of volatile organic chemicals, some systematic monitoring has been instituted to assess the range and variabilities of likely human exposures, particularly in outdoor ambient air, and more recently indoor air as well. Because of the wide use of petroleum and other fossil fuels and the volatility of many of their chemical components, aliphatic and aromatic hydrocarbons have been widely studied both in outdoor and indoor air. Thus for example, Simoneit has shown that aerosols from urbanized areas of the western U.S. contain extractable organic matter composed mainly of petroleum residues (*1*). A National Academy of Sciences report notes that a variety of aromatic and aliphatic hydrocarbons have been detected in indoor air (*2*).

However, there has been increasing attention to low molecular weight, volatile, halogenated compounds because of their wide use, well-known toxicological effects, and awareness of their presence in the environment that results in actual or potential human exposures, such as from contaminated groundwater and improperly managed hazardous waste sites. An example of an evaluation of such exposures to and risks from some of these chemicals is found in a publication of the National Academy of Sciences (*3*) which shows that indoor and outdoor air exposures for many of these chemicals can be as important as those from food and water. Because these and other low molecular weight halogenated organics are potential agents of chronic human disease, the normally low air exposures are of concern and need to be assessed further. The focus of this paper will then be on such compounds in indoor and outdoor air, with particular attention to the chlorinated methanes and ethylenes for which the environmental data base has begun to be developed.

Three separate aspects will be addressed. First will be a discussion of physicochemical characteristics that will affect the movement and interactions of volatile chemicals between gaseous, aerosol and water phases. Second, will be a brief survey of several chlorinated methanes and ethylenes in indoor and primarily outdoor air. Finally, there will a discussion of one particular route of indoor release of volatile chemicals, namely from potable waters used within homes. This will include an evaluation of parameters that will affect human

*Center for Environmental Epidemiology and Department of Industrial Environmental Health Sciences, Graduate School of Public Health, University of Pittsburgh, Pittsburgh, PA 15261.

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exposures from this route, evidence of such releases within homes, and experimental simulation of such emissions.

Physicochemical Aspects

The volatilization of chemicals into air has physicochemical components that affect the likely concentrations that can occur, as well as the fraction that may be found in the particulate as compared to the gaseous state. Of direct interest are the vapor pressures of the pure compound P_A, expressible in such units as mm Hg (torr) or atmospheres. Table 1 shows such values of vapor pressure at 25°C for several anthropogenic organic chemicals, along with their Henry's Law coefficients H, which is the ratio of their air concentrations in equilibrium with those in water. Thus, at equilibrium with P_A (having units of atm) and the water concentration C_W (in units of mole/m^3)

$$P_A = HC_W \quad (1)$$

H is often conveniently estimated as the ratio of the vapor pressure of a pure compound divided by its water solubility (*7*).

Table 1. Henry's Law constant and vapor pressure at 25°C for several anthropogenic chemicals.

Compound	Vapor pressure, mm Hg	H, atm-m^3/mole
Tricholoroethylene	74[a]	9.9×10^{-3}[b]
Benzene[c]	95	5.5×10^{-3}
Aroclor 1248[c]	4.9×10^{-4}	3.5×10^{-3}
Lindane[c]	9.4×10^{-6}	4.9×10^{-7}
DDT[c]	1.0×10^{-7}	3.9×10^{-5}

[a] Data of Dilling et al. (*4*).
[b] Data of Roberts and Dandliker (*5*).
[c] Data of Makay and Leinonen (*6*).

Although there is the temptation to define volatile chemicals of interest simply as those with vapor pressures above a given value, e.g., 1 mm Hg, the H values are also of importance. The larger the H, the greater is the likely human exposure through the air route for a given vapor pressure. A possible reference point in this regard is that value of H which would correspond to equal quantities of the chemical in 20 m^3 of air and 2 L water, the approximate daily amounts taken in by reference man (*8*). On this basis with the use of Eq. (1), the criterion value of H would be 2.5×10^{-6} atm-m^3/mole, which would satisfied by all the compounds in Table 1 except for lindane. As is apparent, compounds with widely different vapor pressures can meet the criterion of H being greater than 2.5×10^{-6}.

The Henry's Law relationship is also relevant to the concentrations of volatile organic chemicals that may be found in rainfall, such as those reported by Kawamura and Kaplan (*9*) as shown in Table 2. Their presence is not unexpected in those ambient atmospheres containing these compounds in the gaseous state, since the large surface to mass ratio of the water droplets in precipitation encourages the rapid equilibration between the air and water phases. For example, for tetrachloroethylene using an H value of 1.5×10^{-2} atm-m^3/mole at 25°C (*5*) and the rain concentration of 21 ng/L shown in Table 2, one calculates a corresponding air equilibrium concentration of 7200 ng/m^3, which is not substantially different from the range of means of 2000 to 4000 ng/m^3 for several U.S. cities reported by Singh et al. (*10*).

Table 2. Concentrations of several volatile organic chemicals in Los Angeles rainwater.[a]

Compound	Concn, ng/L
1,2-Dichloroethylene	230
Chloroform	250
1,1,1-Trichloroethylene	69
Tetrachloroethylene	21
Toluene	76

[a] Data of Kawamura and Kaplan (*9*).

Another essential physicochemical interaction is that discussed by Junge, namely, the likely sorption of a volatile component onto aerosols (*11*). Using the well-known Langmuir absorption relationship, he calculated the expected absorbed fraction of volatile chemicals as a function of aerosol concentration and vapor pressure of the pure compound. His calculations showed that as the aerosol surface concentration per volume of air θ increases, so does the fraction ϕ of the volatile chemical absorbed onto the aerosol surface. Using a typical upper limit of θ for urban aerosols of 2×10^{-5}, his relationships indicate that only for vapor pressures greater than 10^{-4} mm Hg will ϕ be small and most of the volatile chemical exist in the unsorbed vapor form. As shown in Table 1, the vapor pressures of lindane and DDT are well below this value, while those of trichloroethylene, benzene, and Aroclor 1248 are well above it.

Junge's analysis does not address specifically the expected increase in sorbability onto aerosols containing organic matter. Nevertheless, this concept is an important one. Thus both high values of H and the vapor pressure of the pure compound are necessary for volatile organics to constitute an important source of exposure in the gaseous state. If sorbed onto aerosols, such compounds can also clearly constitute a health hazard. However, their behavior and potential for removal are different than when in the gaseous state. Such controls on the distribution of volatile organic chemicals between the gaseous state, aerosols, and water need to be understood and considered in assessing human exposures to such chemicals in air.

Field Surveys

Two recent field surveys of outdoor air concentrations in U.S. cities are of particular interest in that they include low molecular weight volatile halogenated organic chemicals. One is by Singh et al. which was a

study in 1980–1981 of seven widely scattered cities (*10*). The second investigated three urban areas in New Jersey in 1981–1982, as reported by Harkov et al. (*12*).

A summary of some of the results of these studies is shown in Table 3 for several chlorinated methanes and ethylenes. For comparison, surface level background concentrations are shown, based primarily on data from a Pacific marine site at 40° N. In every instance, with the exception of the New Jersey values for carbon tetrachloride, the mean concentrations are substantially above background, and quite variable among the cities. The maximum values reported by Singh et al. (*10*) and shown in Table 3 range from 100 to 1000 times the background concentrations. Also, the variability among cities is substantial. For seven U.S. cities (Table 3), the range of the means varied from a factor of about two for tetrachloroethylene to as high as seventeen for dichloromethane.

Diurnal variations can be quite pronounced as shown for trichloroethylene (TCE) in Houston and Denver by Singh et al. (*10*). For both cities the concentrations were considerably above the remote area background concentration of 15 ppt (parts per trillion), and the late night concentrations were distinctly higher than those in the daytime. The mean diurnal concentrations were as high as 400 to 500 ppt. Knowledge of such variations are of importance in assessing human exposures and in undertaking a sampling program.

Similarly, substantial daily variations were also found for TCE in three cities in New Jersey in the summer of 1981, as reported by Harkov et al. (*12*). They noted that 90% of TCE is used for degreasing and cleaning metals, and that peak concentrations are likely to reflect such regional meteorological events as stagnation. They found that such peak concentrations of volatile organics in the summer of 1981 were as much as two to ten times the seasonal average, and that these occurred during periods of stagnation and a high pressure system. A comparison of the geometric mean concentrations in ppb for four halogenated organic chemicals during these episodic periods with the means for the full six week sampling period in 1981 is shown in Table 4 for the three New Jersey cities. In Newark and Elizabeth all the mean halogenated organic chemical concentrations were higher in the episodic periods.

It is also of interest to compare indoor and outdoor concentrations for these chlorinated volatile organics, although data collected systematically for this purpose are not generally available. Such indoor air measurements have been made by Harsch and Rassmussen, as reported by the National Academy of Sciences (*3*). The range of these reported concentrations are shown in Table 5 for dichloromethane, chloroform, and carbon tetrachloride along with the corresponding outdoor air concentrations from Table 3. The range of the indoor concentrations is considerable and can be substantially greater than background. Nevertheless, it is typically within the general range encompassed by the outdoor air measurements. However, it should be emphasized that the comparative data do not reflect a study of spe-

Table 3. Outdoor air concentrations for several U.S. cities for selected chlorinated organics.

	Air concentrations, ppt (V/V)						
	Seven U.S. cities, 1980–1981[a]				New Jersey, 1981[b]		
Compound	Range of means	Max.	Background	Staten Island	Newark	Eliz.	Camden
Dichloromethane	390–6,760	57,000	50	1,610	350	230	720
Chloroform	81–900	5,100	20	150	60	190	40
Carbon tetrachloride	130–400	2,900	135	310	10	10	10
Trichloroethylene	96–230	2,500	15	170	500	270	210
Tetrachloroethylene	290–590	7,600	50	290	450	310	240

[a] Data of Singh et al. (*10*), arithmetic means, one week period.
[b] Data of Harkov et al. (*12*), geometric means, six weeks in summer.

Table 4. Ambient air concentrations of chlorinated volatile organics in three New Jersey cities in oxidant episode and other periods.[a]

	Chlorinated volatile organics, ppb					
	Newark		Elizabeth		Camden	
Compound	EP[b]	SWG[c]	EP[b]	SWG[c]	EP[b]	SWG[c]
Vinylidene chloride	0.59	0.38	0.78	0.35	0.31	0.36
Methylene chloride	0.56	0.35	0.27	0.23	0.48	0.72
Trichloroethylene	0.69	0.50	0.70	0.27	0.23	0.21
Perchloroethylene	0.56	0.45	0.70	0.31	0.26	0.24

[a] Data of Harkov et al. (*12*).
[b] Episode periods associated with regional oxidant (7/19–21, 8/3–5, 8/10–13), geometric means.
[c] Six weeks geometric means.

Table 5. Comparison of some indoor and outdoor air concentrations for chlorinated methanes.

Compound	Air concentrations, ppt (V/V)			
	Outdoor air[a]			Indoor air[b] (range)
	Range of means	Max.	Background	
Dichloromethane	390–6,760	57,000	50	170–14,000
Chloroform	81–900	5,100	20	14–730
Carbon tetrachloride	130–400	2,900	135	140–460

[a] Data of Singh et al. (*10*), arithmetic means, seven cities.
[b] NAS data (*3*).

cific indoor sites relative to their corresponding outdoor locations.

Indoor Volatilization from Water

Volatilization of chemicals from indoor uses of water can be a substantial source of human exposure as discussed by Prichard and Gesell for radon-222 (*13*). As shown in Table 6, indoor water uses other than for drinking and kitchen constitute the greatest sources of radon release, the efficiency of transfer for radon from water to air varying from 30 to 90%, the weighted mean being 50%. These data indicate that the volatilization from indoor water uses can be quite variable and substantial.

Wadden and Scheff (*14*) have discussed the various models used to estimate expected indoor pollutant concentrations, including one- and multidimensional models, as well as empirical models based on statistical evaluation of concurrent indoor and outdoor air concentrations and other relevant terms. The single compartment model has been widely used and can be greatly simplified to include a mixing factor k to take into account incomplete air mixing, along with the rate of air exchange with the outdoors, q.

On using such a model for indoor volatilization of chemicals from water, the expected steady-state air concentration C_A can thus be calculated simply from and is proportional to a source term S for airborne or volatile materials with units of mass per hour, such that

$$C_A = S/(kq) \quad (2)$$

For k and q values of 0.15 and 338 m^3/hr, respectively, C_A becomes equal to $0.02S$. Where the source is the water used indoors, S is simply the product $30C_W$, where 30 L/hr is a typical indoor water use for a family of four and C_W is the concentration in the water of the chemical of interest. It is assumed the latter is completely volatilized. Substituting these relationships into Eq. (2) one obtains

$$C_A = 0.6C_W \quad (3)$$

Thus, the expected air concentration is simply directly proportional to the water concentration, the proportionality constant being 0.6 L/m^3. The corresponding dimensionless constant is 6×10^{-4}. This number is useful to compare with the dimensionless H values, which for trichloroethylene is about 0.4. The significance is that when the H value is substantially above the indoor air source term constant, the tendency to reach equilibrium is displaced greatly in the direction of essentially complete volatilization. Whether this in fact will occur will depend to a great extent on the time of exposure to air and the surface area of the water use.

The next question is that of the relative exposures to a volatile pollutant from the ingestion and inhalation routes. Taking adult man as an example, one can use a daily respiratory volume of 20 m^3 and a daily water intake of as much as 2 L or as little as 0.15 L, which encompass a likely range that can be encountered (*8*). The daily ingestion from water of a volatile constituent is then calculated simply by multiplying the concentration in water (mg/L) by either 2 or 0.015. Using Eq. (3) to estimate C_A from the volatilization process, the daily intake from air is the product of 20 times C_A, the air concentration. The resulting relationships are presented in Table 7 for the generalized water concentration of a constituent C_W and a specific example of the latter, namely, 0.01 mg/L. It is apparent that exposure

Table 6. Radon release from domestic use of water containing radon, 1000 pCi/L.[a]

Use	Daily consumption, L	Transfer efficiency, %	Radon liberated, pCi
Showers	150	63	94,500
Tub baths	150	47	70,500
Toilet	365	30	109,500
Laundry	130	90	117,000
Dishwasher	55	90	49,500
Drinking and kitchen	30	30	9,000
Cleaning	10	90	9,000
Total	890		459,000

[a] Data of Prichard and Gesell (*13*).

Table 7. Estimated indoor inhalation and ingestion exposures for an adult male from volatile chemicals in water using a one-compartment indoor air quality model.

Water concn, mg/L	Uptake, mg/day		
	Air	2 L intake	0.15 L intake
C_W	$12C_W$	$2C_W$	$0.15C_W$
0.01	0.12	0.02	0.0015

from the air route is substantially greater than that from water ingestion using this single-compartment air model and the assumptions stated above. If a daily ingestion of 2 L is assumed, the air uptake is higher by a factor of 6. This ratio becomes much larger, namely 80, if the smaller estimate of 150 mL per day is used for water ingestion. One should be cautious, however, in using these ratios as more than an example of the relative uptakes that can occur via the drinking water and air inhalation routes within the home. Nevertheless, they do indicate the possibility of substantially greater air exposures to volatilized constituents from water used within the home and, therefore, the need to consider this route of exposure in assessing possible health effects from such contaminants in potable water supplies.

Couch and Andelman (*15*) investigated the possible volatilization of trichloroethylene (TCE) into indoor air within buildings in a small community using individual wells obtaining water from an aquifier measured to contain about 40 mg TCE
. By using a continuous real-time monitor with an infrared detector, measurements were taken in closed rooms in two homes and a small municipal building on one day in July 1983. Prior to turning on water in bathrooms, no TCE could be detected in the indoor air above the detection limit for the instrument, namely 0.5 mg/m^3. However, TCE was readily detected in the bathrooms with water running. The air concentration levels increased with time as shown in Table 8, as expected. In home B, the highest concentration measured after 17 min of the shower running was 81 mg/m^3, approximately one-third of the American Conference of Government Industrial Hygienists (ACGIH) time-weighted threshold limit value of 270 mg/m^3 for the work environment (*16*). To estimate the possible dose within these homes from these shower inhalation exposures, one can take a value of about 40 mg/m^3 as the average air concentration during the shower period. Assuming 1 hr per week in the shower and an air breathing volume of 1.2 m^3 in that hour, this would correspond to a potential intake of 48 mg TCE/week (neglecting any question of the fraction of the dose that is absorbed). If one also assumes an ingestion of the contaminated water of 150 mL/day (2.1 L/week), the water dose if 42 mg TCE/week, indicating that shower air and ingestion doses are comparable. One would also expect an increased air dose from exposure elsewhere in the home as a result of TCE volatilizing from other water uses.

Table 8. Indoor air concentrations of tricholoroethylene (TCE) in buildings using well water containing TCE, ca. 40 mg/L.[a]

Location	Time	Concn, mg/m^3
Municipal bldg.		
Ladies rest room, water running	12:20 PM	35
Home A		
Kitchen	1:34 PM	ND[b]
Bathroom,	1:53	32
shower on	1:56	48
	2:01	67
	2:03	72
Home B		
Bathroom,	2:20 PM	ND
upstairs,	2:27	19
shower on	2:30	35
	2:33	40
	2:37	81
Bathroom,	3:08 PM	ND
downstairs,	3:17	64
shower on	3:24	67

[a] Data of Couch and Andelman (*15*).
[b] ND = not detected above detection limit of 0.5 mg/m^3.

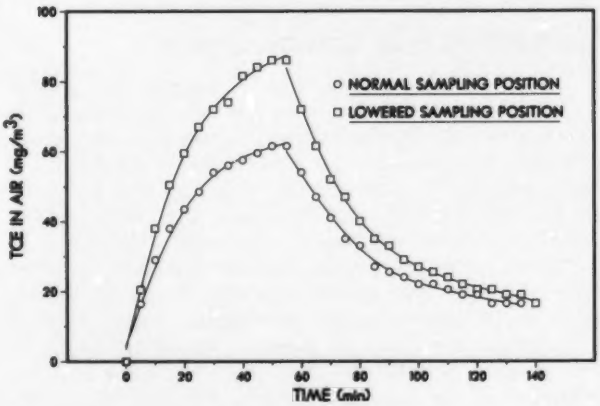

FIGURE 1. TCE in air withdrawn from shower.

Experiments have been undertaken by Couch and Andelman (*15*) to simulate the air exposures that can be encountered in showers using water contaminated with volatile chemicals, initially trichloroethylene (TCE). Some early results are given in Figure 1, which shows the build-up of TCE in air withdrawn from the shower at two sampling positions. When steady state was reached after about 55 min the injection of TCE was discontinued, and the subsequent decay was also monitored. The air withdrawal rate was 0.05 shower volumes/min; the shower volume, 0.1 m^3; the water flow rate, 0.2 L/min; TCE water concentration, 3.8 mg/L; and the water temperature, 23°C.

Although the TCE is injected continuously, as it volatilizes it is expected and was found that the air concentration did level off to a steady state, the concentration at which the rate of volatilization equals the rate of withdrawal by the controlled air flow. There is evidence for nonuniform air mixing, as shown in Figure 1 by the higher steady-state air concentration with the lowered sampling position. It was calculated that if the TCE were to volatilize completely, the predicted uniformly mixed steady-state concentration would have been 152 mg TCE/m^3 (0.76 mg TCE/min input to the shower divided by 0.005 L air/min withdrawn from shower), substantially higher than the measured values

of 62 and 86. The lower values could indicate incomplete volatilization, but incomplete mixing, as noted above, is likely to have accounted for a good part of the difference.

It is apparent from such experimental and field studies that both high point-source exposures, as well as more generalized high indoor air levels can arise by volatilization of chemicals from indoor water uses. Since many groundwater supplies are known to be contaminated by volatile chlorinated organics, this route of exposure should be considered in assessing sources of indoor air pollutants, as well as in the development of regulations to limit such exposures from public water supplies.

Summary and Conclusions

Low molecular weight volatile organic chemicals of anthropogenic origin, including chlorinated compounds, are widely distributed in urban ambient air at concentrations substantially above background levels in remote areas. Not unexpectedly they can also be found at similar concentrations in indoor air. Although the outdoor air concentrations are typically in the range of parts per billion and lower, there are considerable diurnal and longer time variations which should be considered in estimating human exposures. Many of these compounds are likely or suspect human mutagens or carcinogens, so that continuing surveillance and assessment of the human exposures and possible health effects is prudent.

The indoor and outdoor air concentrations of volatile chemicals in the gaseous state are greatly influenced by physicochemical properties, especially solubility in water, vapor pressure of the pure compounds, their ability to sorb onto air particulates, and the rates at which they can volatilize from aqueous solution. Such properties will affect not only their ultimate concentrations in ambient outdoor air, but also indoor air exposures. Lower aqueous solubility will increase the likelihood of their being found in the gaseous state, as will high vapor pressure. Even with volatilization, reduced vapor pressure will increase the likelihood of their sorption onto air particulates.

Volatilization of chemicals from indoor water uses is of growing interest and concern, particularly as water supplies become increasingly contaminated. Resulting indoor air exposures and the factors that can influence them need to be further characterized. Although the regulation of toxic chemicals in potable water supplies has focused traditionally on direct ingestion, the volatilization and inhalation from other much greater volume indoor uses of water should be considered, with focus also on the large exposures that can result from bathing and showering.

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